PATENT SPECIFICATION

(11) 1 535 832

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(21) Application No. 2177/76 (22) Filed 20 Jan. 1976
(31) Convention Application No. 542529
(32) Filed 20 Jan. 1975 in
(33) United States of America (US)
(44) Complete Specification published 13 Dec. 1978
(51) INT CL² B01D 13/00
(52) Index at acceptance
B1X 6A1 6B4 6B6 6D1 6F4 6F7 6FX 6HX 6J5 6K

(19)

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(54) PRESSURE DRIVEN MEMBRANE PROCESSING APPARATUS

(71) We, EASTMAN KODAK COMPANY, a Company organized under the Laws of the State of New Jersey, United States of America of 343 State Street, Rochester, New York 14650, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to the separation of a liquid from, dissolved or suspended solid material in a feedstream, e.g. separation of a solute from the solvent by passing said solvent preferentially through a semipermeable membrane. More particularly, this invention relates to separation by means of ultrafiltration, dialysis, reverse osmosis, and similar processes, i.e., pressure-driven membrane processes, to a method of cleaning the membranes utilized in such processes, and to apparatus for effecting such cleaning.

While this invention is applicable to a wide variety of separation operations, it is especially beneficial to filtration by a reverse osmosis process, and as a matter of convenience will be described principally in terms of such a process.

The term "solution" will be used hereinafter to mean a true solution wherein a solid solute is dissolved in a solvent, and the term "suspension" will be used to mean a feed liquid comprising a solvent having suspended undissolved solid solute therein.

In reverse osmosis processes generally, a separation of solvent from solute is achieved by passing the feed stream, for example an industrial waste stream, into contact with a semipermeable membrane, the feed stream being under sufficient pressure to overcome the osmotic pressure across the membrane and force the solvent (e.g. water) to pass through the membrane and out of the solution, leaving the remaining rejected solution more concentrated. This type of process is designated "reverse osmosis" (RO) because the movement of solvent through the membrane is in a direction opposite to that which would normally be brought about by virtue of concentration differences. The most commonly used RO membrane is cellulose acetate as described in detail in the patents and publications mentioned hereinafter.

In such processes, as in the various other membrane-separation processes referred to above, there is a tendency for the efficiency, e.g. permability (and rejection) of the membrane, to be impaired by so-called "fouling". The exact mechanism is unknown, but all types of reverse osmosis membranes and all membrane unit configurations are subject to fouling. Membrane fouling has been found to arise from a number of causes, including precipitation of salts, suspended and/or particulate matter, and attachment of organic macromolecules. Many liquid wastes have substantial quantities of suspended solids therein, and these wastes have not been easily treated by membrane processes because of fouling.

Many processes have been proposed to control membrane fouling and to restore fouled membranes to a high level of efficiency. United States Patent No. 3,491,021 teaches the use of reciprocating angular acceleration of the membrane and it support relative to the solution-suspension to prevent accumulation of filter cake on the membrane surface. United States Patent No; 3,425,562 discloses the association of a movable element with the membrane to increase flow velocity and turbulence adjacent to the membrane surface to dislodge particles trapped thereon. French Patent No. 2,082,051 provides for a cleaning process for liquid filters where filtration is effected by means of pre-coated elements. To effect the removal of pre-coat, which tends to

clog the element, the pressure vessel is opened, the element withdrawn, and a gas, such as compressed air or an inert gas, is projected onto the deposits formed on the element while rotating it, causing the deposits to detach therefrom. United States Patent No. 3,630,360 teaches the utilization of a pressure reversal from one side to the other of the filter every 15 to 30 second to dislodge the solids layer from the filter in a system wherein a flexible filter element, concave under filtering conditions and convex under non-filtering conditions, is employed. British Patent No. 1,276,524 teaches the removal and replacement of exhausted precoat particles. A cartridge, which is vertically positioned in a filter tank, is cleaned by draining influent from the tank until it is below the cartridge, and gradually refilling the tank by passing a mixture of gas and waste liquid into and through the cartridge in reverse flow, to gradually fill the tank with wash liquid at least to the top of the cartridge. The waste liquid is then drained off while the air flow is continued.

Other partially satisfactory proposals include the creation of turbulence such as by high feed liquid velocity, the presence of a mechanical agitator, or by membrane configuration, the addition of detergents and/or acids, back flushing, and depressurisation. In some situations, feed pre-treatment, i.e. filtering, adding reducing agents, adding precipitation inhibitors, controlling pH to prevent inorganic precipitates, and passing feed through activated charcoal columns, is effective to prevent fouling, but will not reduce fouling in a membrane already contaminated.

We have found that membranes in a pressure-driven membrane process, having surfaces which have become fouled as feed liquid flows in contact therewith, can be cleaned and rejuvenated by the periodic purging of the fouled membrane surfaces with large volumes of fast moving gas, while maintaining the membrane in a wet condition.

In accordance with the present invention, there is provided a method for operating a pressure driven membrane processing apparatus comprising the steps of passing a flow of feed liquid containing a dissolved or suspended solid into a processing unit having a semi-permeable membrane wall, the feed liquid being in contact with a first side of the membrane wall, removing a liquid permeate of reduced solid content at a second side of the membrane wall, opposite the first side and removing a liquid concentrate of increased solid content from the feed liquid, and removing accumulated fouling matter from the first side of the membrane wall by a gas purge cycle comprising the steps of

a) interrupting the flow of feed liquid to the processing unit

b) expelling the residual feed liquid from the processing unit by means of a stream of gas injected into the processing unit and

c) discontinuing the stream of gas to the processing unit, whereafter the flow of feed liquid is reintroduced into the processing unit.

Where the accumulated fouling matter is such that the membrane cannot be completely cleaned by the gas purge alone, the method of the present invention also provides a secondary purge cycle which may be used in combination with the gas purge cycle, to clean the membrane surface. Two forms of secondary purge cycle may be used. Firstly, the secondary purge cycle may comprise the additional step of introducing into the processing unit, after discontinuing the stream of gas, a flow of feed liquid at a pressure below that required to drive the feed liquid through the membrane wall and thereafter increasing the pressure of the feed liquid to a pressure above that required to drive the feed liquid through the membrane wall.

Alternatively, the secondary purge cycle may comprise the additional steps of introducing into the processing unit, after discontinuing the stream of gas a) a flow of wash liquid at a pressure below that required to drive the wash liquid through the membrane wall b) discontinuing the flow of wash liquid into the processing unit c) reintroducing the flow of feed liquid into the processing unit at a pressure above that required to drive the feed liquid through the membrane wall.

Frequent repetition of the gas purge cycle in combination with the second purge cycle may be undertaken until the membrane surface is cleaned.

The period between such gas purge cycles, when they are followed by the secondary purge cycle, is of much greater duration than in the case where the gas purge cycle is used alone (the frequency of such cycles therefore being low). It is again pointed out that the method of this invention is applicable to most pressure-driven membrane processes, e.g. to those pressure-driven membrane processes that would not be adversely affected by the large volumes of gas required.

The flow of purging gas is directed parallel to the active surface of the membrane, along the path taken by the feed liquid. The direction of the purging gas relative to the feed liquid flow direction can either be in the direction of such feed liquid flow, or in the opposite direction, i.e. in a back-flushing mode.

The duration of the gas purge cycle, and the frequency with which the apparatus is to be purged, are dependent on the variables of the apparatus. These variables include membrane area, number of and configuration of discrete membrane units, whether the membranes are already fouled when the method is first applied, and the type of feed being treated and the fouling it produces, which in turn determines whether or not it is necessary to include the secondary purge cycle in order to clean the membrane. It has been found that each system requires a minimum volume and pressure of gas to clean and prevent fouling of the membrane. This minimum volume and pressure of gas is ascertainable through a trial and error method, but the duration (the length of the period of injecting gas into contact with the membrane element) of the gas purge cycle may be reduced by the use of greater gas pressure, which will provide the necessary volume of gas in a shorter time period. Such reduction may be desirable to keep the apparatus "on stream" for the maximum possible time without fouling material building up on the surface of the membrane.

The method of this invention is most efficiently incorporated into a particular apparatus by determining the minimum purge time, both in frequency and duration. Where the gas purge cycle is employed alone the minimum gas purge cycle repetition frequency may be determined by purging the apparatus at gradually increasing intervals until the flux across the membrane decreases, indicating the onset of membrane fouling. Operation at a slightly shorter interval than that at which flux decrease was noted will give maximum "on stream" time. The duration of the gas purge cycle may be decreased, by increasing the gas pressure to provide a larger volume of gas in a shorter time period, such being constrained by equipment limitations and the economics of the process.

As the duration of the gas purge cycle is dependent on the pressure of the gas used during such a cycle, this method of determining the minimum frequency and duration of the gas purge cycle may be repeated until the maximum efficiency of the apparatus is established. After a period of operating the apparatus, the frequency and duration of the gas purge cycle may be reassessed to keep process efficiency at its maximum.

When the secondary purge cycle comprising the low pressure liquid feed flow is utilised in combination with the gas purge cycle little filtration takes place during this secondary purge cycle because the liquid pressure is kept low; therefore, for maximum efficiency the apparatus should be maintained in the filtering mode for as long as possible between purging cycles. The duration of the secondary purge cycles, however, must also be kept as short as possible, as this results in undesired non-filtering time. Determination of the minimum secondary purge repetition frequency may be begun by arbitrarily picking a set period of time for keeping the apparatus in the filtering mode. The pressure within the apparatus is then lowered to below that required for filtration, and the secondary purge cycle utilizing repeated reintroduction of low pressure feed liquid flow after gas purging is employed until the membrane surfaces are cleaned. Completion of the cleaning process is determined by raising the pressure within the apparatus into the filtration range and ascertaining if the flux decrease across the membrane due to the fouling has been reversed and the flux restored to its initial level.

The duration of the combination of the gas purge cycle and the secondary purge cycle may be decreased by determining the optimal duration and frequency of the gas purge cycle, as previously described for the gas purge cycle above. In this manner, the shortest possible non-filtering time is realised.

After a period of operating the apparatus the frequency and duration of the secondary purge cycle may be reassessed to keep process efficiency at its maximum. Incorporation of the method of this invention into apparatus containing already fouled membranes will require more frequently purging with gas until the membrane surfaces are cleaned, whereupon the frequency and duration of purging may be reduced by application of the hereinbefore described procedures.

The gas purging method herein described is applicable both to a processor comprising a single semi-permeable membrane unit and to a processor comprising a plurality of pressure-driven semi-permeable membrane units connected in series or parallel arrangement. Multi-element filtration systems can be cleaned utilizing the method of this invention either by purging the entire system at once, or by sequentially purging the several membrane units, one at a time. Purging the entire system at once has the drawback of requiring a much greater volume of air at a higher pressure than that required for the sequential method.

In the accompanying drawings:—

Fig. 1 is a schematic representation of a single membrane, reverse osmosis filtration apparatus constructed according to one form of the present invention using timed

motorized valves for performing the gas purging method;

Fig. 2 shows a portion of the apparatus represented in Fig. 1, but employing a check valve for controlling air flow instead of a timed motorized valve;

Fig. 3 is a schematic representation of a modification of the single membrane, reverse osmosis filtration apparatus shown in Fig. 1, providing for recirculation of feed;

Fig. 4 is a schematic representation of a parallel connected multi-membrane reverse osmosis filtration apparatus for performing the gas purging method of the invention;

Fig. 5 is a schematic representation of a single membrane reverse osmosis filtration apparatus incorporating still another embodiment of the apparatus of the invention, providing for recirculation of concentrate;

Fig. 6 is a graph showing rejection of solids in concentrate plotted against days of operation while processing sewage plant secondary effluent feed, showing the beneficial effect of gas purging by the method of this invention without a secondary purge cycle; and

Fig. 7 is a graph showing permeate flow rate changes plotted against days of operation while processing sewage plant secondary effluent feed, showing the improved effect of gas purging by the method of this invention, without a secondary purge cycle, versus three methods of the prior art.

Referring to Fig. 1, there is shown a pressure-driven membrane unit of the reverse osmosis type, utilizing a single semi-permeable membrane unit 11 which includes a gas purge apparatus constructed in accordance with one form of the present invention. The apparatus comprises a reservoir 12 containing a liquid solution which is pumped by pump 13 through two-way valve 15 into a feed line 16 and then into the membrane unit 11. A valve 17 is located in a gas line 18 which joins the feed line 16 downstream from two-way valve 15. When the two-way valve 15 is open and valve 17 is closed, feed liquid passes into the membrane unit 11 and is separated by operation of the membrane into a concentrate steam and a permeate stream. The concentrate stream containing an increased concentration of solute flows into concentrate stream line 19 and passes out through throttle valve 20. Throttle valve 20 serves, via back pressure, to regulate the pressure within the apparatus as provided by the pump, and which may be adjusted to increase or decrease pressure in the apparatus, such as to the desired level for reverse osmosis filtration, which is greater than the osmotic pressure of the solution and generally is 150 to 700 psi or greater (all pressures are gauge pressures herein). If the pump and system are designed to provide the necessary operating pressure for filtration, throttle valve 20 is necessary only to provide a low pressure path for a purging gas controlled by valve 17 during a purge cycle, as hereinafter described. Two-way valve 15 and valve 17 are connected to timed valve actuating motor T1, and throttle valve 20 is connected to timed valve actuating motor T2, but valves 15 and 20 can be operated manually if desired.

Membrane configurations for membrane unit 11 which can be utilized in this sytem include spirally-wrapped reverse osmosis permeators, as shown in United States Patent Nos. 3,367,504; 3,386,583 and 3,397,790, and large tube permeators, as disclosed by Walters in *Mechanical Engineering*, April 1968, pp. 104—110.

The operation of the apparatus shown in Fig. 1 is as follows: In normal "on stream" filtering operation, feed liquid is pumped by pump 13 at a pressure above the osmotic pressure thereof from reservoir 12 through two-way valve 15, which is in an open position with respect to feed line 16, into the membrane unit 11 where it is separated into concentrate and permeate, the concentrate passing through throttle valve 20 which is partially open to concentrate stream line 19. Throttle valve 20 is adjusted to provide back pressure, thereby providing the pressure required in the apparatus for filtration.

At a predetermined time, a purging cycle is carried out as follows. Two-way valve 15 is closed by timed valve motor T1 to stop feed liquid input to membrane unit 11 although the pump 13 continues to operate. As two-way valve 15 closes, throttle valve 20 is rapidly opened to its full extent via the action of timed valve motor T2, so as to completely open concentrate stream line 19, thereby allowing the pressure in membrane unit 11 to drop to atmospheric pressure. Coincidental to this further opening of throttle valve 20, timed valve motor T1 slowly opens valve 17 in gas line 18 such that the pressure in the membrane unit 11 is effectively atmospheric before the valve 17 begins to admit purging gas. The gas, such as compressed air, flows through gas line 18 and feed line 16 into the membrane unit 11. This gas flows along the active surface of the membrane, disturbing and loosening the fouling substances on the surface of the membrane, and blowing substantially all of the feed liquid remaining in the membrane unit 11 through throttle valve 20 and out of the apparatus. Timed valve motor T1 now reopens two-way valve 15 and feed liquid is

pumped back into the reverse osmosis membrane unit 11 at the operating pressure of the apparatus. Simultaneously, timed valve motor T1 closes valve 17 stopping the flow of gas into membrane unit 11, and timed valve motor T2 returns throttle valve 20 to its original partially open position. This purging cycle is repeated at selected time intervals which, as hereinbefore described, have been found to be most efficient with respect to preventing membrane fouling and maintaining high "on stream" time. A typical application where the membrane unit is being used to treat photographic emulsion wash water containing particulates, requires 5 seconds of gas purging with a one minute liquid feed period between such purges. A two minute liquid feed period between such purges, under similar conditions, requires 12 seconds of gas purging.

Referring to Fig. 3, there is shown a modified apparatus in which two-way valve 15 is replaced by a three-way valve 15′, to which is connected a bypass line 21. Three-way valve 15′ when actuated by timed valve motor T1, closes feed line 16 while opening bypass line 21 such that the pump recycles feed liquid back to reservoir 12. Upon reactuation by timed valve motor T1, three-way valve 15′ reconnects pump 13 to feed line 16 and closes by-pass line 21. Otherwise, the apparatus and its operation are identical to those described in connection with Fig. 1.

When a secondary purge cycle is to be imposed on the purging cycle described, the throttle valve 20 and its timed valve motor T2 are so designed and adjusted that upon completion of the purging cycle, throttle valve 20 is not returned to the original throttling position which assures high liquid filtration pressure, but remains instead at a more open intermediate position so that, via back pressure, the feed liquid, when reintroduced into membrane unit 11, is at a pressure below or equal to the pressure of the gas, generally from 30 to 100 psi. Thus, the feed liquid flows along the fouled membrane surface without permeating the membrane, washing away fouling material which has been loosened by the gas. The gas purge and secondary purge cycles are repeated as often as needed until the membrane surfaces are clean, then timed valve motor T2 restores throttle valve 20 to its original throttling position to develop the desired high back pressure (e.g. 150 psi or greater) required for the feed liquid to permeate the membrane.

The gas purge cycle and the secondary purge cycle, are repeated at time intervals which have been found to be most efficient with respect to preventing membrane fouling and maintaining high "on stream" time. A typical application of this method using a secondary purge cycle, where the membrane unit is being used to treat photographic emulsion wash water containing particulates, requires 60 minutes of secondary type purging, with a 23 hour period of high pressure feed liquid flow before purging is again necessary.

The choice of which of the hereinbefore set forth purge cycles of this invention to utilize, i.e. the gas purge cycle followed by high operating pressure feed liquid flow, or the gas purge cycle followed by the secondary purge cycle using low pressure liquid flow is governed primarily by the type of feed that the permeator is treating. Three basic feed types may be encountered: non-sticky particulate feeds such as untreated or raw waste from photographic processes; organic chemical-containing feed, such as emulsion wash water from photographic processes; and feed which produces sticky particulate or organic fouling, such as a gelatin-containing waste stream which has been previously treated with cannibalizing bacteria.

It has been found that gas purging followed immediately by feed liquid at high operating pressure effectively restores membranes exposed to fouling from non-sticky particulate and organic chemical feeds, but is of reduced effectiveness when applied to membranes fouled by sticky particulate or organic feed, such as bacteria-containing gelatin waste feed. It is hypothesized that this difficulty is due to the deposition of bacteria bodies and various degradation products on the membrane surface during liquid filtration. The gas purging is able to loosen and partially lift these materials from the membrane surface, but the reintroduction of feed liquid flow at high filtration operating pressure "plasters" the lifted portions of the materials back onto the membrane surface, resulting in virtually no removal of the loosened materials. In this situation, the addition of the low pressure feed liquid flow purging method is effective. The reintroduction of feed liquid flow at low pressure, it is hypothesized, does not cause reattachment of the loosened or lifted materials so that continued repeated secondary purge cycles will remove them and clean the membrane surface. It has also been found that the low pressure feed liquid flow purging effectively removes sticky particulate and organic fouling where bacteria bodies and degradation products are not present, although a hypothesis as to why the method is effective in such situations is not readily reached.

Thus, when applying this invention to regenerate membranes in a membrane unit filtration system, the gas purge cycle followed by operating pressure feed liquid flow may first be tried. If this provides insufficient membrane cleaning, the secondary purge cycle using low pressure feed liquid flow may then be applied.

All of the procedures described above in connection with Figs. 1 and 2 also apply when the single reverse osmosis membrane unit 11 is replaced with two or more such units connected together in series. The permeate can be removed from each unit individually, but the concentrate and purging gas pass through the successive units and then out through a concentrate line at the end of the series. Alternatively, the series of units can be positioned within a single pressure casing, and the permeate discharge conduits can be connected together in series, with the combined permeate flowing out of the last unit in the series, as described in U.S. Patent 3,417,870.

Referring to Fig. 2, a simpler form of the apparatus uses a check valve 17' in the gas line 18 which is designed to remain closed at high operating pressure, instead of the valve 17. When cleaning of membrane unit 11 is required, valve 15" is closed (manually or automatically) and the pressure in feed line 16 drops from the operating pressure (e.g., 150 psi) down to a lower pressure such as 90 psi at which the closed check valve 17' is designed to open automatically and start the flow of gas into the membrane unit 11. When valve 15" is reopened, high pressure feed liquid enters the membrane unit 11, and check valve 17' automatically closes. This apparatus can operate with or without a throttle valve in concentrate line, providing the required back pressure develops.

With reference to Figure 4, there is represented a parallel arrangement of membrane units 25 and 27, set up so as to comprise a multi-unit process system. With such apparatus, two alternative purging procedures are possible. First, both membrane units 25 and 27 are purged with gas at the same time and second, the units 25 and 27 are purged at different times.

In the first procedure, the purging cycle begins with three-way valves 29 and 31 closing off feed lines 33 and 35 into membranes units 25 and 27, the pumps 37 and 39 (or single pump when used) continuing to operate. As three-way valves 29 and 31 close with respect to feed lines 33 and 35, partially open two way valves 41 and 43 are rapidly opened via the action of timed valve motor T3, so as to completely open concentrate lines 45 and 47, thereby allowing the pressure in membrane units 25 and 27 to drop to atmospheric pressure. Coincidental to the opening of two-way valves 41 and 43, timed valve motors T4 and T5 slowly open valves 49 and 51 in gas lines 53 and 55, such that the membrane units are effectively at atmospheric pressure before the valves 49 and 51 begin to admit gas. Gas passes into feed lines 33 and 35 and thence into membrane units 25 and 27 to clean the membrane surfaces as previously described. Timed valve motor T4 then reopens three-way valves 29 and 31 with respect to feed lines 33 and 35 and feed liquid is pumped into membrane units 25 and 27. Simultaneously, timed valve motors T4 and T5 close valves 49 and 51 terminating the flow of gas, and timed valve motor T3 returns two-way valves 41 and 43 to their original partly open throttling positions, such that the pressure within the apparatus rises to that required for filtration.

The secondary purge cycle described in connection with Fig. 1, using low pressure feed liquid flow, may also be applied to the apparatus shown in Fig. 4 so as to purge both membrane units 25 and 27 at the same time. The procedure is as that previously described, save that upon completion of the gas purge, timed valve motor T3 does not return two-way valves 41 and 43 to the position such that they provide filtration back pressure, but returns them to a more open intermediate position so that the feed liquid is at a pressure near or equal to the pressure of the purging gas. The purging cycles are repeated until the membrane surfaces are clean. At that point, three-way valves 29 and 31 are reopened with respect to feed lines 33 and 35 and, simultaneously valves 49 and 51 are closed and two-way valves 41 and 43 are returned to their original throttling positions, enabling the pressure within the apparatus to rise above the osmotic pressure of the feed liquid.

In a modification of Fig. 4, the pumps continue to pump feed liquid through by-pass lines back to the reservoir 12. This is accomplished as previously described in connection with Fig. 3 but using two bypass lines.

Alternatively, the purging cycle can be arranged so that membrane unit 25 is purged while membrane unit 27 is "on stream". Membrane unit 27 is then purged while membrane unit 25 is "on stream". The timed valve motors T3, T4 and T5 are programmed to set valves 31, 51 and 43 for feed liquid treatment and valves 29, 49 and 41 for the purging cycle (with or without imposition of the secondary purge cycle) and vice versa. During purging of membrane unit 25, valve 29 is open with respect to line 54 to enable feed liquid to flow through valve 31 into membrane unit

27, and vice versa. Normally, both membrane units 25 and 27 are simultaneously on stream as the duration of the purging cycle is short compared to the duration of the feed liquid treatment period.

A further embodiment of the apparatus of this invention is provided where the configuration of the membrane system is such that exhausting the purging gas into the concentrate line as previously described may lead to problems in such lines, such as the formation of a gas lock. The apparatus as shown in Fig. 5 provides bypass means and a valve which allows the application of the gas purging method of this invention without resulting gas locks in concentrate lines.

Referring to Figure 5, the operation of the apparatus is exactly as in Fig. 1 except that the concentrate passes from membrane unit 11 through a three way valve 56, which is in a closed position with respect to bypass line 57 and open with respect to concentrate stream line 19. The concentrate flows through partially open throttle valve 20 which is positioned so that it serves a throttling function to provide back pressure, thereby providing the desired apparatus pressure for filtration.

At a predetermined time, e.g., at the beginning of the purging cycle, valves 15, 17 and 20 are operated as described in connection with Fig. 1 and/or Fig. 3. As two way valve 15 closes with respect to feed line 16, three-way valve 56 by the action of timed valve motor T6, closes rapidly with respect to concentrate stream line 19 and opens with respect to bypass line 57, thereby connecting the membrane unit 11 to the reservoir 12 via the bypass line 57, and allowing the pressure in membrane unit 11 to drop to atmospheric pressure. Thus, when the purging gas flows through membrane unit 11, the residual liquid remaining in the membrane unit 11, together with fouling material dislodged from the membrane surface returns to reservoir 12 by way of bypass line 57. Conversely, when the valves 15, 17 and 20 return to their original positions, three way valve 56 is actuated to again close off bypass line 57 and open concentrate stream line 19.

Instead of operating the apparatus of Fig. 5 only with a gas purge cycle followed by high pressure feed liquid flow, a secondary purge cycle can be employed wherein lower pressure liquid flow is repeatedly alternated with gas purging before high pressure feed liquid flow is resumed. This has been previously described in detail with reference to Fig. 1.

Furthermore, a bypass line such as 57 and a three way valve such as 56 can be used in both the series connected and parallel connected systems described previously.

The mechanism by which the gas purging of this invention cleans the membrane surfaces is not completely understood, but it is hypothesized that it arises from a combination of factors. When the gas flows into the membrane unit it drives the feed liquid remaining therein before it, probably giving rise to a turbulent, bubbling effect which acts on the surface of the membrane to loosen fouling substances. In a cylindrical spirally wrapped membrane unit the gas also causes an expansion of the membrane as it forces its way into the feed stream path, giving rise to a flexing action in the membrane. When substantially all of the residual feed liquid has been expelled from the membrane unit the gas flow is terminated, and the feed liquid stream reintroduced. It is essential that the flow of gas be terminated while the membrane is still in a wet condition, as, if the membrane is dried, its filtering characteristics are damaged or even destroyed. The reintroduced high pressure feed liquid flow surges into the cartridge, now driving the residual gas before it, producing turbulence which acts further to loosen fouling substances from the membrane surface. The reintroduction of the feed liquid flow also causes an expansion in the cylindrical spirally wrapped membrane unit, again giving rise to flexing forces which act on the surface of the membrane. It is this combination of forces, due to the sequential removal of feed, introduction of gas, and reintroduction of feed that is believed to result in the cleaning of the membrane surfaces.

Preferably, the gas used in the method of this invention is air, which may be provided by known compression equipment. It has been found that the volume of air usually required is such that a holding tank or accumulator providing storage for a large quantitiy of compressed air may be necessary. The volume required will depend on the particular system involved. While air is preferred, any other gas such as nitrogen or argon may be used which will not deleteriously interact with either the active surface of the membrane or the feed liquid which remains therein during the purging cycle.

The methods and apparaus of this invention may be used to clean and prevent fouling in processing systems which process a wide range of feed liquids. Such apparatus is effective in preventing fouling where feed liquids comprise, for example, industrial sewage, effluent waste water from paper production operations, emulsion wash

90 psi = 0.6 MPa

water from photographic film coating operations, sugar solutions, and fruit juices.

The following examples illustrate the application of the methods and apparatus of this invention to pressure-driven membrane processes.

Example 1

A prefiltered solution of secondary effluent from an industrial sewage treatment plant was continuously processed, with recycle of concentrate, utilizing cellulose acetate reverse osmosis membranes in a spirally wrapped unit such as described in U.S. Patents 3,386,583 and 3,417,870; operating conditions are set forth in Table 1. Figures 6 and 7 show the effects of fouling on membrane rejection and flux, for the unit used, during several days of operation.

As shown at points X, Y and Z in Fig. 7 attempts were made following the prior art to reduce the fouling effects by flushing the apparatus with aqueous solutions of the named commercial detergents)"Soilax", a neutral pH detergent; "Impact", a high pH detergent; and "Lime-Away", a phosphoric acid and surfactant solution, all trademarks of Economics Laboratories, Inc.). The beneficial effects of such prior art cleaning were minimal compared to the excellent improvement shown for gas purging by the method of the present invention at point F.

At 18 days of operation, the invention was employed in the following manner: the spirally wrapped membrane unit was subjected to 30 minutes of alternating passage of water and air, 15 second periods at 90 psi for each. Figures 6 and 7 show that the air purging method of the invention resulted in marked improvements in both permeate flux rate and solids rejection by the previously fouled membrane upon resumption of feed liquid flow at a pressure above the osmotic pressure. Rejection is defined as the amount of solids held back by the membrane, and is calculated by subtracting the weight of solids in the permeate from the weight of solids in the feed, and then dividing the difference by the weight of solids in the feed and multiplying by 100.

Conditions appear in Table 1 below:

TABLE 1

Pump:	Multistage Centrifugal
Permeator	Osmonics Spiral Wound "SEPA 97"
RO Membrane:	Cellulose Acetate
Feed Liquid Pressure:	150 psi in; 125 psi out
Feed Flow Rate:	5 gallons per minute
Temperature (average):	75°F
Prefilter:	15 micron Fulflo filter (Fulflo is a trademark)
Air and water injection:	90 psi

Example 2

Wash water from photographic emulsion manufacture, containing 10 ppm of silver as silver compounds, as well as other heavy metal compounds, was treated by passing it through a Gulf ROGA90 cellulose acetate spiral permeator, 27 sq. ft. area, at a pressure of 400 psi and a temperature of 45—60°F., with recycle of concentrate. A 250 gallon sample was concentrated to 5 gallons, 98% of the water having passed through the membrane as permeate and being available for reuse. The limited solubility of the heavy metals caused them to precipitate during concentration. This precipitate has a content of 50,000 ppm of recoverable silver.

The apparatus shown in Fig. 5 was used, but with recycle of feed to the reservoir during air purging as shown in Fig. 3. A cycle was employed of 2 minutes on-stream with feed liquid flow, followed by purging with compressed air at psi for 15 seconds, and then resuming 400 psi feed liquid flow.

Flux through the membrane remained high, and the particulate content of the concentrate remained high throughout the run, indicating that fouling was minimized. Without air injection, early fouling was expected, based on previous experience.

Example 3

The apparatus of Example 2 was operated in the same way for 1000 hours. Only a slight flux decrease from .34 to .31 gal/minute was observed over the entire 1000 hour period.

Under the same conditions, but without periodic air injection, 200 gallons of photographic emulsion wash water were concentrated, with recycle of concentrate, to observe the fouling rate. After 500 hours of operation the flux dropped to .13 gal/minute from the initial flux of .31 gal/minute.

Example 4

A prefiltered aqueous feed liquid containing a major proportion of organics such as enzyme - degraded gelatin, and a minor proportion of compounds of silver and other metals was pumped through a

reverse osmosis membrane unit (Osmonics 97 Spiral Permeator) in apparatus similar to that shown in Fig. 5, but with provision for recycle of feed liquid to the reservoir during air injection by replacing the valve 15 of Fig. 5 by the three-way valve 15′ as shown in Fig. 3.

The feed liquid was at a pressure of 200 psi and a temperature of 95°F. After 70 hours on stream the flux had dropped from the initial 325 ml/minute to 150 ml/minute, at which time the three-way control valve 15′ was closed automatically, valve 17 was automatically opened to admit air at a pressure of 80 psi to membrane unit 11, and three-way valve 56 was adjusted for recycle of feed liquid. Periodically, three-way valve 15′ was opened automatically, throttle valve 20 was automatically adjusted for the desired back pressure, and three-way valve 56 was returned to its original position, to supply feed liquid at a pressure of 30 psi and temperature of 130°F., while air valve 17 was closed automatically. This cycle was continued for 6 hours, with air and low pressure feed liquid each flowing for 15 second periods. Then three-way valve 15′ was reopened, and throttle valve 20 was readjusted to supply high pressure feed liquid to the membrane unit. It was found that the flux was 350 ml/minute, indicating that the loosened fouling material had been removed from the membrane surfaces and permeability restored.

The operations described above were generally performed on a laboratory scale with recycle of liquid concentrate back to the feed liquid reservoir for repeated passage through the permeator. In commercial operations the feed liquid would generally be passed through the apparatus only once, as is possible when employing a series of permeators, and there would be no recycle of concentrate.

Several timed valve motors have been shown and described for actuating the several valves. It should understood that the valves can be operated in the proper sequence by individual pneumatic, hydraulic, or electrical motors, all of which are connected to and controlled by any known type of timer mechanism (not shown).

In Example 4, with 15 second periods for each half of the low pressure secondary purging cycle, the length of the periods only relates to the specific membrane unit. In general, the air is injected for a period slightly longer than that required to expel all residual liquid from the apparatus (this may vary from 10 seconds to 60 seconds or more, depending on the liquid capacity of the unit or units). The low pressure liquid flows for a period long enough to refill the cavity in the membrane unit and wash out the material loosened by the air (this too will vary from 10 to 60 seconds or more, as with air).

The washfluid can be any suitable liquid, and may contain chemical adjuvants as required for particular operations. For example, the liquid can contain acid or alkali for pH adjustment, or surfactants to improve its detergent action. In this case the flow of wash fluid is discontinued before reintroducing the feed liquid to the apparatus at the operating pressure.

WHAT WE CLAIM IS:—

1. A method for operating a pressure driven membrane processing apparatus comprising the steps of passing a flow of feed liquid containing a dissolved or suspended solid into a processing unit having a semi-permeable membrane wall, said feed liquid being in contact with a first side of the membrane wall, removing a liquid permeate of reduced solid content at a second side of the membrane wall opposite said first side and removing a liquid concentrate of increased solid content from the feed liquid, and removing accumulated fouling matter from said first side of the membrane wall by the additional steps of interrupting the flow of feed liquid to the processing unit, expelling the residual feed liquid from the processing unit by means of a stream of gas injected into the processing unit and discontinuing the stream of gas to the processing unit, whereafter the flow of feed liquid is reintroduced into the processing unit.

2. A method according to Claim 1 wherein removal of accumulated fouling matter from the first side of the membrane wall includes the additional step of introducing into the processing unit, after discontinuing the stream of gas, a flow of feed liquid at a pressure below that required to drive the feed liquid through the membrane wall, and thereafter increasing the pressure of the feed liquid to a pressure above that required to drive the feed liquid through the membrane wall.

3. A method according to Claim 1 wherein the removal of accumulated fouling matter from the first side of the membrane wall includes the additional steps of introducing into the processing unit after discontinuing the stream of gas, a flow of wash liquid at a pressure below that required to drive the wash liquid through the membrane wall and discontinuing the flow of wash liquid to the processing unit, whereafter the flow of feed liquid is reintroduced into the processing unit at a pressure above that required to drive the feed liquid through the membrane wall.

4. A method according to Claim 2 or 3,

wherein the steps of introducing a stream of gas, discontinuing said stream of gas and introducing a liquid at a pressure below that required to drive the liquid through the membrane wall are carried out two or more times prior to the reintroduction of the feed liquid at a pressure above that required to drive the feed liquid through the membrane wall.

5. A method according to any one of the preceding Claims wherein the pressure of the feed liquid in the apparatus is between 150 and 700 psi.

6. A method according to any one of the preceding Claims wherein the gas is air.

7. A method according to any one of the preceding Claims wherein the flow of feed liquid, the flow of gas and the reintroduced flow of feed liquid is regulated automatically on a timed cycle.

8. A method according to any one of the preceding Claims wherein the flow of gas is injected into the processing unit in the same direction as the flow of feed liquid.

9. A method according to any one of the preceding Claims 1 to 7 wherein the flow of gas is injected into the processing unit in the opposite direction to the flow of feed liquid.

10. A method according to any one of the preceding Claims as appendant to Claim 2, wherein the flow of feed liquid which is reintroduced into the processing unit at a pressure below that required to drive the feed liquid through the membrane wall is at a pressure of 30 to 100 psi.

11. A method according to any one of the preceding Claims wherein the removed liquid concentrate is the feed liquid for another processing apparatus connected in series to the first processing apparatus.

12. A method according to any one of the preceding Claims wherein the feed liquid flows simultaneously into at least two processing apparatus arranged in parallel and that the entire sequence of removing accumulated fouling matter from one apparatus is performed completely before it is performed on a second apparatus.

13. A method according to Claim 1, substantially as hereinbefore described.

14. Pressure-driven membrane processing apparatus comprising a reservoir of feed liquid, a membrane unit having a semi-permeable membrane wall for effecting processing of the feed liquid, a first feed line interconnecting the reservoir with the membrane unit, a pump for pumping the feed liquid from the reservoir through the first feed line, separate second and third feed lines for removing concentrate and permeate streams from the membrane unit, respectively, a first control valve in the first feed line down stream from the pump and operable to start and stop the flow of feed liquid to the membrane unit, a fourth feed line connected into the first feed line downstream of the first control valve a source of gas connected to the fourth feed line and a second control valve in the fourth feed line, wherein the second control valve is normally closed when the first control valve is open, and is open when the first control valve is closed, so as to allow gas to flow into and through the first feed line and into the membrane unit.

15. Processing apparatus according to Claim 14 comprising a third control valve in the second feed line located downstream from the membrane unit for controlling back pressure in said unit.

16. Processing apparatus according to Claim 14 or 15 comprising a motor operatively connected to the first and second control valves, and a timer controlling the energizing of said motor on a timed cycle, such that the control valves are operated automatically in a timed sequence.

17. Processing apparatus according to Claim 14, 15 or 16 wherein the first control control valve is a three-way valve, and the processing apparatus also comprises a by pass line connecting the three-way valve to the reservoir so as to allow the recirculation of the feed liquid through the by pass line when the three-way valve is closed with respect to the membrane unit.

18. Processing apparatus according to Claim 14, 15, 16 or 17, comprising first and second membrane units for effecting processing; the first feed line interconnecting the reservoir with the first and second membrane units; the first control valve individually controlling the flow of feed liquid, and the second control valve individually controlling the flow of gas, so that when the first valve is open the second valve is closed.

19. Pressure-driven membrane processing apparatus according to Claim 1 constructed and adapted to operate substantially as herein before described with reference to, and as shown in, the accompanying drawings.

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Agent for the Applicants.

Printed for Her Majesty's Stationery Office, by the Courier Press, Leamington Spa, 1978
Published by The Patent Office, 25 Southampton Buildings, London, WC2A 1AY, from which copies may be obtained.

FIG. 1

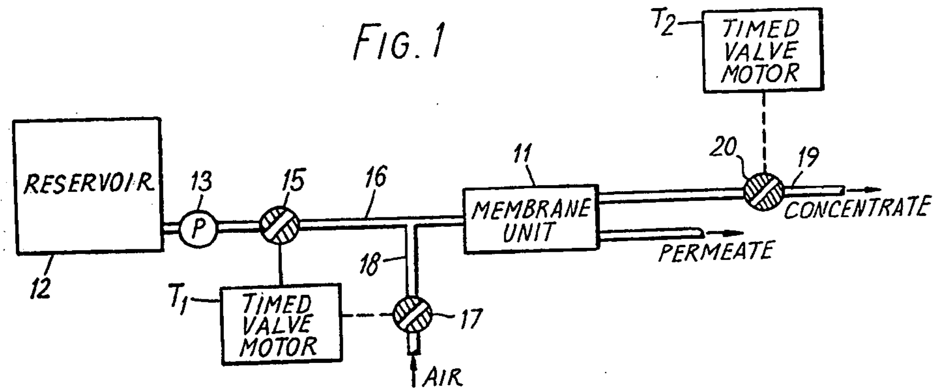


FIG. 2

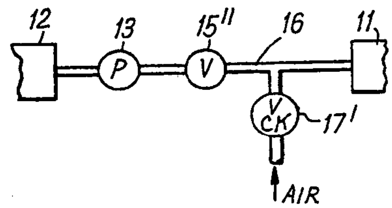


FIG. 3

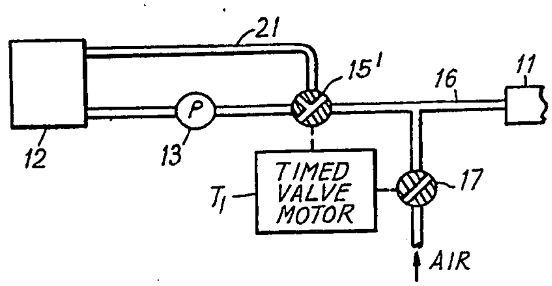
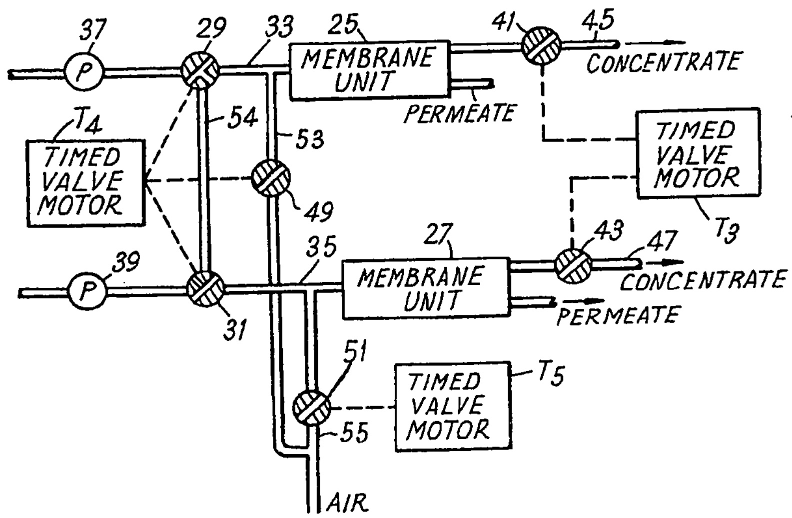


FIG. 4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P
29
33
25
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41
45
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PERMEATE
T4
TIMED VALVE MOTOR
54
53
49
TIMED VALVE MOTOR
T3
27
43
47
35
39
P
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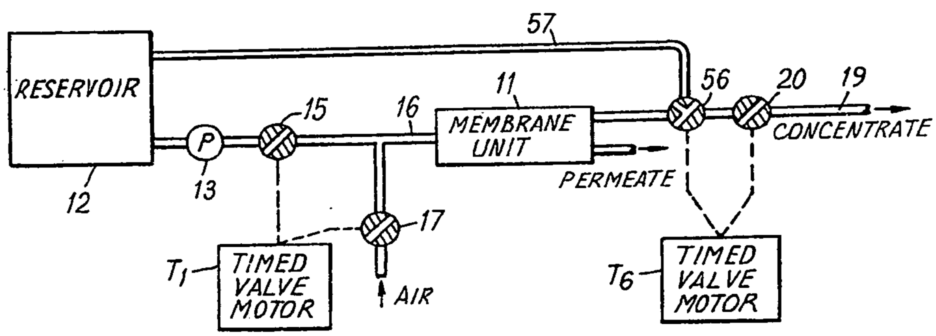
CONCENTRATE
PERMEATE
31
51
TIMED VALVE MOTOR
T5
55
AIR

FIG. 5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16
MEMBRANE UNIT
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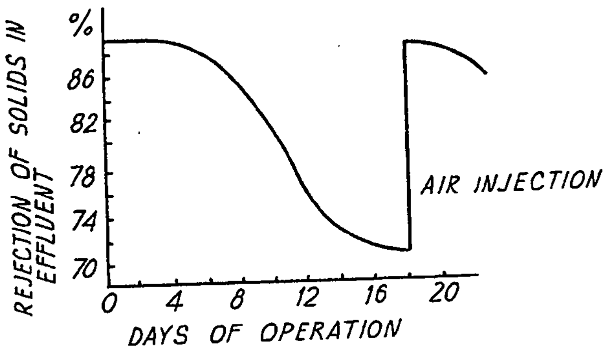
P
PERMEATE
12
13
17
T1
TIMED VALVE MOTOR
AIR
T6
TIMED VALVE MOTOR

FIG. 6


%
86
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REJECTION OF SOLIDS IN EFFLUENT
AIR INJECTION
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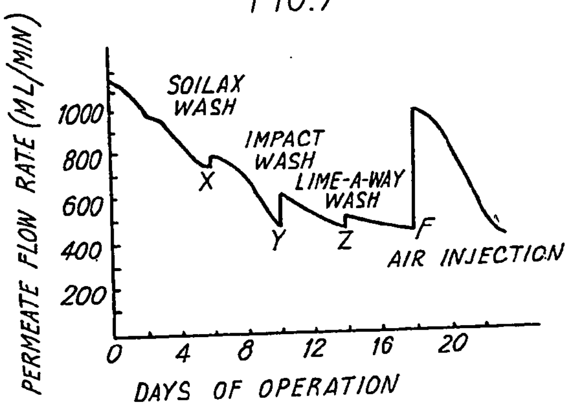
4
8
12
16
20
DAYS OF OPERATION

FIG. 7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200
SOILAX WASH
IMPACT WASH
LIME-A-WAY WASH
X
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Z
F
AIR INJECTION
0
4
8
12
16
20
DAYS OF OPERATION